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Direction of Magnetization in Thin Films

—Molecular-field Theory—

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Abstract

The phase transition of thin films having perpendicular anisotropy at the surface layers and in-plane anisotropy in inner layers is studied by the molecular-field approximation. For a fixed in-plane anisotropy, three ordered phases, i.e., perpendicular ordering, in-plane ordering, and canted-spin states are obtained depending on the strength of the surface anisotropy.

§ 1. Introduction

Recently, the direction of magnetization in thin films consisting of a few layers has been studied intensively [1]—[6]. In particular, the study of perpendicular ordering of very thin films has been of great interest because of its application to magnetic storage technologies. Since thin films are essentially two dimensional, and an isotropic two-dimensional system has no long-range order, the magnetic ordering of thin films is dominated by the presence of anisotropy. Due to the demagnetization field caused by layered structure, films in general have in-plane anisotropy which forces the spins to be parallel to the layers. The surface, however, in certain conditions may have uniaxial perpendicular anisotropy which originates from spin-orbit couplings, and if the perpendicular anisotropy overcome the in-plane anisotropy, the thin film will exhibit perpendicular ordering.

In this report, we propose a simple model which can describe the competition between perpendicular and in-plane anisotropy of a thin ferromagnetic film, and study how the direction of magnetization changes with temperature and film thickness. In § 2 we set up a model Hamiltonian, and use the molecular-field approximation to calculate the magnetization in each layer. In § 3 the free energy and the method to determine the phase diagram are given. In § 4 we perform numerical calculations and discuss the results obtained.

§ 2. Model Hamiltonian and Magnetization

We consider a ferromagnetic thin film of thickness N with perpendicular anisotropy in the surface layers and in-plane anisotropy in the inner layers. To describe such a system, we introduce the Hamiltonian

$$H=-J\sum_{\langle ij,l'j'\rangle}(\xi_{ll'}S^x_{ij}S^x_{l'j'}+S^y_{ij}S^y_{l'j'}+\eta_{ll'}S^z_{ij}S^z_{l'j'}), \tag{1}$$

where J is a measure of the strength of the ferromagnetic exchange coupling, l and l' layer indices, j and j' lattice points in the layers. Anisotropy factoes $\xi_{ll'}$ and $\eta_{ll'}$ represent Ising-like one, and take $\eta_{ll}>1$ and $\xi_{ll}=1$ on the surface layers, while $\eta_{ll}=1$ and $\xi_{ll}>1$ in the inner layers. Therefore, spins in surface layers tend to poing perpendicular (z) direction, while those in inner layers tend to lie in the plane of the film (x direction). Interplane exchange couplings are assumed to be isotropic: $\xi_{l,l\pm1}=\eta_{l,l\pm1}=1$.

In order to determine the stable direction of spins (z'-axis) within a molecular-field approximation, we first rotate x- and z-axes by angle ϕ_l about y-axis,

$$\begin{cases} S^x_{lj}=S^{z'}_{lj}\sin\phi_l+S^{x'}_{lj}\cos\phi_l \\ S^y_{lj}=S^{y'}_{lj} \\ S^z_{lj}=S^{z'}_{lj}\cos\phi_l-S^{x'}_{lj}\sin\phi_l. \end{cases} \tag{2}$$

Then, rewriting the Hamiltonian (1) by the new coordinates and neglecting two-body terms of spin operators, we have a molecular-field Hamiltonian

$$H_M=\frac{1}{2}J\sum_{ij}(E_i\mu_i+K_i\tau_i)$$

$$-J\sum_{ij}[(K_i\sin\phi_i+E_i\cos\phi_i)S_{ij}^{z'}+(K_i\cos\phi_i-E_i\sin\phi_i)S_{ij}^{x'}], \tag{3}$$

where

$$\begin{cases} E_i=4\eta_{ii}\mu_i+\mu_{i-1}+\mu_{i+1} \\ K_i=4\xi_{ii}\tau_i+\tau_{i-1}+\tau_{i+1}. \end{cases} \tag{4}$$

Here we have assumed a simple cubic structure, and μ_0, μ_{N+1}, τ_0, and τ_{N+1} vanish by the boundary condition. The constant term in Eq. (3) should not be discarded if the free energy is of interest. As illustrated in Fig. 1, μ_i and τ_i, which are respectively the vertical and parallel component of the magnetization S_i of the i-th layer, are given by

$$\begin{cases} \mu_i=<S_{ij}^{z}>=S_i\cos\phi_i \\ \tau_i=<S_{ij}^{x}>=S_i\sin\phi_i \\ S_i=<S_{ij}^{z'}>. \end{cases} \tag{5}$$

Since z'-axis is chosen as a quantization axis, the stable direction of spins is determined by vanishing the coefficient of $S_{ij}^{x'}$ in Eq. (3),

$$\tan\phi_i=\frac{K_i}{E_i}=\frac{\tau_i}{\mu_i}, \tag{6}$$

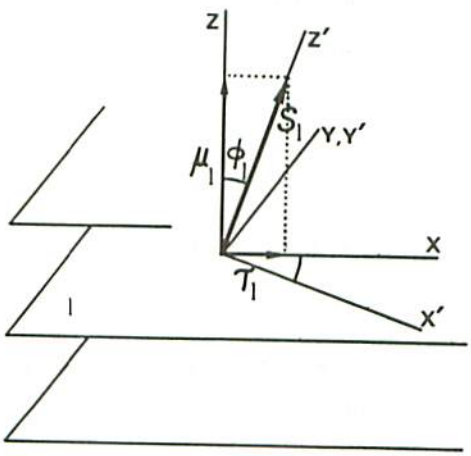

Fig. 1 Illustration of the geometry of the magnetic moment and the coordinate system.

and by substituting this into Eq. (3) the molecular-field Hamiltonian is reduced to

$$H_M=\frac{1}{2}J\sum_{ij}(E_i\mu_i+K_i\tau_i)-2J\sum_{ij}\xi_i S_{ij}^{z'} \tag{7}$$

with

$$\xi_i=\frac{\sqrt{E_i^2+K_i^2}}{2}. \tag{8}$$

If we treat the case with spin-1/2, average magnetization S_i is calculated in the usual way:

$$S_i=\frac{\mathrm{Tr}(S_{ij}^{z'}\hat{\rho})}{Z}=\frac{1}{2}\tanh(J\beta\xi_i), \tag{9}$$

where Z is the partition function

$$Z=\mathrm{Tr}\hat{\rho}=\mathrm{Tr}\ \exp(-\beta H_M), \tag{10}$$

where $\beta=1/(k_B T)$, k_B the Boltzmann constant, T the absolute temperature. By solving $2N$ self-consistent equations obtained from Eqs. (5), (8), and (9), we can determine $2N$ unknowns μ_i and τ_i.

§ 3. Free Energy and Phase Diagram

A set of self-consistent equations given in § 2 will yield four types of solutions corresponding to z-ordering, x-ordering, canted-spin, and paramagnetic states. Which phase is the stable one is determined by comparing the free energy of the four phases. The free energy

$$F=-\beta^{-1}\ln Z \tag{11}$$

is calculated easily as

$$f=\frac{1}{N}\sum_i\left[\frac{J}{2}(E_i\mu_i+K_i\tau_i)-\beta^{-1}\ln\{2\cosh(J\beta\xi_i)\}\right] \tag{12}$$

per atom.

The phase boundary between a z-ordering or x-ordering state and a canted-spin state is determined by the condition

$$E_i\tau_i-K_i\mu_i=0, \tag{13}$$

because this holds in the canted-spin states as found in Eq. (6), and also is consistent with the result of the perturbation expansion of the free energy. The phase boundary

between a z-ordering state and a canted state is given by solving the homogeneous equation

$$(E_l-4\zeta_{ll}\mu_l)\,\tau_l-\mu_l\tau_{l-1}-\mu_l\tau_{l-1}=0, \tag{14}$$

or equivalently,

$$\begin{vmatrix} E_1-4\mu_1 & -\mu_1 & & & \\ -\mu_2 & E_2-4\zeta_b\mu_2 & -\mu_2 & & \\ & \cdot & & & \\ & & \cdot & & \\ & & -\mu_{N-1} & E_{N-1}-4\zeta_b\mu_{N-1} & -\mu_{N-1} \\ & & & -\mu_N & E_N-4\mu_N \end{vmatrix}=0. \tag{15}$$

Here and in the following, anisotropy factors are chosen such that $\eta_{11}=\eta_{NN}=\eta_s$ for surface layers, and $\zeta_{ll}=\zeta_b$ for inner layers $l=2, 3, \cdots, N-1$. As assumed in our model, $\zeta_{11}=\zeta_{NN}=1$ and $\eta_{ll}=1$ for inner layers. The condition which gives the phase boundary between a x-ordering state and a canted-spin state is the same as in Eq. (15) except that E_l, ζ_{ll}, and μ_l are replaced by K_l, η_{ll}, and τ_l, respectively.

The phase boundary between a z-ordering or x-ordering state and a paramagnetic one is of second order and defines the Curie temperature T_c. Near T_c, the right hand side of Eq. (9) can be expanded as a series in powers of μ_l or τ_l. Keeping only the linear term, we have homogeneous equations

$$4T_c\mu_l=E_l, \tag{16}$$

and

$$4T_c\tau_l=K_l, \tag{17}$$

respectively for the z-ordering and x-ordering states. T_c is determined by setting the determinant of the coefficient matrix in Eqs. (16) and (17) to zero.

§ 4. Results and Discussions

For specified values of in-plane anisotropy ζ_b and film thickness N, we can map the four phases in $(\tilde{T}, \eta_s)$ plane, where $\tilde{T}=k_BT/J$ and η_s being the perpendicular anisotropy at both surfaces. In Fig. 2, we illustrate the phase diagrams for $\zeta_b=1.1$ and $N=3, 4, 5, 6$, where X, Z, C, and P designate respectively the x-ordering, z-ordering, canted-spin, and paramagnetic phases.

From Fig. 2, we can find a general trend of spin arrangement at low temperatures. For small η_s, spins order in plane. When η_s exceeds a certain critical value η_{sx}, the in-plane ordering turns into a canted-spin arrangement. For thin films, the canted arrangement turns into a perpendicular-ordering state at $\eta_s=\eta_{sz}$. The value of η_{sz}

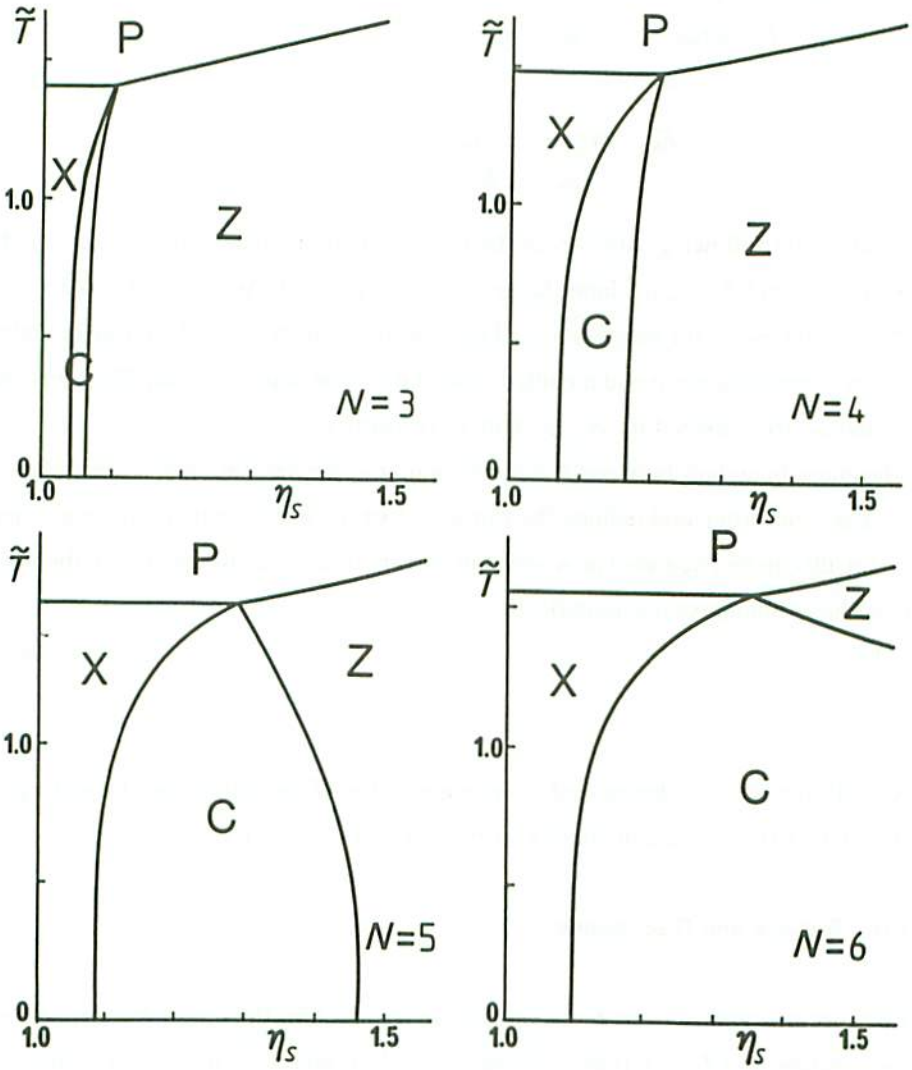

Fig.2 $(\tilde{T}, \eta_s)$ phase diagram for film thickness $N=3,4,5,6$, and $\eta_b=1.1$. X, Z, C, and P designate x-ordering, z-ordering, canted-spin, and paramagnetic states, respectively.

depends very sensitively on N, and for thick films no perpendicular ordering occurs. We can explain these features as follows: At low temperatures, a stable spin arrangement is determined mainly by which state has the lowest energy. If the surface is isotropic ($\eta_s=1$), spins on the surface layer point the same direction as that of spins in inner layers (x direction). Therefore, the x-ordering state still remains as the most stable one provided that η_s is sufficiently small. Since the exchange interaction allows a continuous turning of direction of each spin, as η_s increases the competition between the perpendicular and in-plane anisotropy produces a continuous turning of the magnetization direction from in-plane to perpendicular orientation, so that canted-spin arrangement appears. The phase X and C can exist for any N. The z-ordering state, however, appears only for small N. As was shown in Ref. [7], the effect of the surface is restricted to within a few layers from the surface. Therefore, for thick films, there exist two surface regions and the bulk between them. Even when the spins on the surface almost point z direction, the effect cannot extend to the bulk, and then the bulk spins can almost lie in parallel to the layers. In contrast, for thin films the Ising-like anisotropy in z direction on both surfaces enhances each other to force all the spins in the film to point z direction. In fact, at $T=0$, $\eta_{sz}=1.063$ for $N=3$, which is less than ζ_b ($=1.1$), whereas the corresponding value becomes larger than the ζ_b for N larger than 4.

As the temperature increases, the entropy term contributes to the minimum condition of the free energy. As was shown in Ref. [7], at high temperatures, the layers with strong anisotropy have dominant effect, and the average values of spins become very small on the layers with weak anisotropy. Therefore, near T_c, x-ordering or z-ordering state remains to be stable irrespective of the thickness of the film.

Recently, Pappas *et. al.* [3] found that the magnitude of the perpendicular surface-anisotropy depends on the temperature. It is large at low temperatures and decreases as the temperature increases. As a consequence, very thin films with perpendicular anisotropy show the perpendicular ordering at low temperatures, and then turns into in-plane ordering as the temperature increases. Also, they found signs of canted-spin arrangement. Though the dipole interactions are not included in our model, we can explain qualitatively the behavior of the temperature dependence of direction of magnetization in thin films. Another interesting feature in their study is a finding of the

complete loss of magnetization in the transition region. If we treat Eq. (1) using the equation of motion method, the lowest excitation of the collective mode would become soft at the phase boundary between X and C, and Z and C. In view of the low dimensionality of thin films, we will find a considerable reduction of spins around the boundary. In the molecular-field approximation used in this work, this interesting feature was not taken into account.

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